

Creep failures in heterogeneous materials

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We present creep experiments on fiber composite materials with controlled heterogeneity. Recorded strain rates and acoustic emission rates exhibit a power law relaxation in the primary creep regime (Andrade law) followed by a power law acceleration up to rupture over up to four decades in time. We discover that the failure time is proportional to the duration of the primary creep regime, showing the interplay between the two regimes and offering a method of rupture prediction. These experimental results are rationalized by a mean-field model of representative elements with nonlinear visco-elastic rheology and with a large heterogeneity of strengths.

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The damage and fracture of materials are technologically of enormous interest due to their economic and human cost. Failure of composite systems is particularly important in naval, aeronautics and space industry. Despite considerable experimental [1, 2, 3] and theoretical work [4, 5, 6] on fracture, many questions have not been answered yet. Recently, statistical physicists have shown the existence of a power law acceleration of acoustic emissions announcing the global failure of heterogeneous materials [2, 3], similar to the critical behavior of out-of-equilibrium phase transition [6], offering a way to predict material failure [2].

This letter presents creep experiments on composite materials, which we explain using a simple model of representative elements, in the framework of fiber bundles models. Creep is the progressive deformation of a material under constant load at a given temperature. Three creep regimes are usually observed. During the primary regime, the strain rate decays as a power law with time following the application of the stress (Andrade law) [7]. The secondary regime describes a quasi-constant deformation rate, which evolves towards the tertiary creep regime, if the stress and the temperature are high enough, during which the strain rate accelerates up to rupture.

The experiments are carried out on cross ply glass/polyester composite materials and on Sheet Molding Compound (SMC) composites. Two type of cross angle ply laminates are fabricated, denoted $[\pm 62^\circ]$ and $[90^\circ/35^\circ]$, where the angles measure the directions of the glass fibers with respect to the loading direction, with a fibre volume fraction of 75%. The SMC composites consist of a combination of polyester resin, calcium carbonate filler, thermoplastic additive and random oriented short glass fibers, in the form of a

sheet. The relatively low fibre volume fraction, about 30%, and the uncontrolled filler and reinforcement distribution during processing lead to a more heterogeneous structure for the SMC compared to the cross ply composites. The $[\pm 62^\circ]$ and $[90^\circ/35^\circ]$ specimens have dimensions $14 \times 100 \times 2 \text{ mm}^3$. The SMC samples are in the form of 120 mm barbell with 3 mm tickness. All specimens are subjected to a constant stress s and temperature T (below the glass transition of the matrix), which were fixed to $s = 15 \text{ MPa}$ and $T = 60^\circ\text{C}$ for the $[\pm 62^\circ]$ specimens, $s = 22 \text{ MPa}$ and $T = 60^\circ\text{C}$ for the $[90^\circ/35^\circ]$ specimens, and $s = 48 \text{ MPa}$ and $T = 100^\circ\text{C}$ for the SMC. The creep tensile tests were performed using a servo-hydraulic mechanical testing system. Constant tensile load was applied and the resulting strain and acoustic emissions were recorded until final rupture. Acoustic emissions (AE) is a standard technique to monitor the evolution of damage in composites, due to matrix cracks, fiber matrix debonding, fiber breaks, and delaminations [9]. We used a Mistras data acquisition system by Physical Acoustics Corporation with 2 resonant sensors (200 kHz - 1 MHz).

Normal primary creep transients followed by secondary and tertiary creep were observed for almost all samples, both for the strain rate (Fig. 1) and for the AE rate (Fig. 2). The decrease of the strain rate and EA rate in the primary creep regime can be described by Andrade's law [7] $de/dt \sim t^{-p}$, with an exponent p in the range 0.2-1.4 for the 15 samples tested [8]. The crossover for small times is probably due to the fact that the stress progressively increases up to about 10 sec after the start of the experiment. A quasi-constant strain rate (steady-state or secondary creep) is observed over an important part of the total creep time, followed by an increase of the creep rate up to failure in the tertiary

creep regime. Creep strains at fracture are around 40% for angle cross ply composites and $\approx 4\%$ for the SMC. The acceleration of the strain rate before failure is well fitted by a power-law singularity $de/dt \sim (t_c - t)^{-p'}$ with p' in the range 0.3-1.1 depending on the sample [8]. The critical time t_c determined from the fit of the data with a power-law is close to the observed failure time. Our experiments confirm over large time scales covering up to four orders of magnitude in time previous announcement of power laws in the tertiary creep regime, which were established over more limited time scales [3]. We also obtain the same temporal evolution for the AE energy rate, with larger fluctuations for the energy rate than for the event rate due to the existence of a power-law distribution of AE energies. We found no qualitative differences between the behavior of the $[\pm 62^\circ]$ and $[90^\circ/35^\circ]$ cross ply composites. The values of p and p' are on average a little larger for the SMC than for the cross ply composites, possibly due to the larger heterogeneity of the SMC or to the different values of the applied stress.

There is a huge variability of the failure time from one sample to another one, for the same applied stress, as shown in Fig. 3. This figure shows that the transition time t_m between the primary creep regime and the tertiary regime, measured by the minimum of the strain rate, is proportional to the rupture time $t_m \approx 2/3 t_r$. We also found a negative correlation between the Andrade exponent p and the rupture time t_r [8]. These observations shows that damage in the primary regime impacts on its subsequence evolution in the secondary and tertiary regimes. This suggests a way to predict the failure time from the observation of the strain rate or AE rate during the primary and secondary creep regimes, before the acceleration of the damage leading to rupture.

Creep observations have been modelled in terms of visco-elastic fibers, with deterministic dynamics and quenched disorder [10]. This model reproduces a power law singularity of the strain rate before failure with $p' = 1/2$ in the case of a uniform distribution of strengths [10] but does not explain Andrade's law for the primary creep. Here, we start from the model of [10] and enrich it with a more realistic rheology and heterogeneity, in order to account simultaneously for Andrade's law in the primary creep and for the power-law singularity of the strain rate before failure. We view a composite system as made of a large set of representative elements (RE), each element comprising many fibers with their interstitial matrix. The applied load is shared democratically between all RE. This assumption has been shown to be a good approximation of the elastic load sharing for sufficiently heterogeneous materials [13]. Each RE is modelled

as a non-linear Eyring dashpot [11] in parallel with a linear spring of stiffness E . The Eyring rheology, which is standard for fiber composites, consists at the microscopic level in adapting to the matrix rheology the theory of reaction rates describing processes activated by crossing potential barriers. A given RE fails when its elongation/deformation e reaches a threshold. The rupture thresholds are distributed according to the cumulative distribution $P(e)$ given by $P(e) = 1 - (e_{01}/(e + e_{02}))^\mu$, where e_{01} and e_{02} are two constants with $e_{01} \leq e_{02}$. The fraction $1 - (e_{01}/e_{02})^\mu$ breaks as soon as the stress is applied. The power-law distribution $P(e)$ for large e is motivated by the large distribution of failure times for the same applied stress (Fig. 3). The exponent $\mu > 1$ controls the amplitude of the frozen heterogeneity of the RE strengths.

The equation controlling the deformation $e(t)$ of each surviving RE is

$$\frac{de}{dt} = K \sinh \left(\frac{\beta s}{1 - P(e)} - \beta E e \right) \quad (1)$$

with the initial condition $e(t=0) = 0$. The fraction of unbroken RE is $1 - P(e)$ and $s/(1 - P(e))$ is the stress applied on each unbroken RE.

The system defined by (1) is stable (no global rupture) if the differential equation (1) has a stationary solution $de/dt = 0$ with $e > 0$, i.e., if the equation $((e + e_{02})/e_{01})^\mu = (E/s)e$ has a non-trivial solution. This defines a threshold s^* below which the strain converges asymptotically to a constant and above which de/dt grows without up to rupture.

In the primary regime $e \ll e_{02}$ thus $(e + e_{02})^\mu \approx e_{02}^\mu (1 + \mu e/e_{02})$. If the stress on the dashpot is small, we can replace \sinh by $\exp/2$. With these approximations, the differential equation (1) has the solution

$$\frac{de}{dt} = \frac{K}{2e^{-\beta s} \left(\frac{e_{02}}{e_{01}} \right)^\mu + t K \beta \left(E - \frac{\mu s}{e_{02}} \left(\frac{e_{02}}{e_{01}} \right)^\mu \right)} \cdot \quad (2)$$

Expression (2) predicts that, if the stress is not too large, de/dt is of the Andrade form $\sim t^{-p}$, with an exponent $p = 1$ at early times. For larger s , the strain rate starts to accelerate as soon as the load is applied. Note that the observation of Andrade's power-law creep in this model does not involve any failure of RE and is thus independent of the choice of the distribution of rupture thresholds $P(e)$.

In the tertiary creep regime, we can neglect e_{02} compared with e . Close to failure, for large e , the linear term Ee is small compared with $s/(1 - P(e)) = \frac{s}{e_{01}^\mu} (e + e_{02})^\mu$ if $\mu > 1$. This leads to the equation

$$\frac{de}{dt} \approx \frac{K}{2} \exp \left(\frac{\beta s e^\mu}{e_{01}^\mu} \right). \quad (3)$$

Its solution is, to leading logarithmic order,

$$\frac{de}{dt} = \frac{A}{\mu} [-\ln(t_c - t)]^{\frac{1}{\mu}-1} \frac{1}{t_c - t}, \quad (4)$$

where $A = e_{01}(\beta s)^{-1/\mu}$. Contrary to the primary regime, the heterogeneity of the rupture threshold is an essential ingredient for the power-law singularity before failure, but the leading power-law term with $p' = 1$ in (4) does not depend on the exponent μ characterizing heterogeneity. The acceleration toward failure is due to the positive feedback effect of broken RE, which increases the stress and strain on the unbroken RE leading to the global failure of the system.

Figure 4 shows the numerical solution of equation (1) together with the approximate analytical solutions (2) in the primary creep and (4) close to failure, for different values of the applied stress s . In the primary creep regime close to the rupture threshold $s \approx s^*$, we observe numerically an apparent exponent $p < 1$, smaller than predicted by (2), which can explain the values of p found experimentally [8]. For a stress $s \gg s^*$, the strain rate accelerates immediately when the load is applied. For $s < s^*$, the p -value decreases in the model between 1 and 0 as the applied stress increases. The duration of the primary creep also decreases with s . The model thus explains the correlation found experimentally between the p -value and the failure time [8].

In the tertiary regime, for $s \gg s^*$, we find numerically that expression (3) is a good approximation very close to failure $t \approx t_c$. But for $s \approx s^*$, there is a crossover further from failure with an apparent exponent $p' = 0.9$. This simple model thus reproduces both power-laws in the primary and tertiary creep regimes, with an apparent exponent $p \leq 1$ for the primary creep, and with $p' = 1$ for the tertiary regime, except for a crossover with an apparent exponent p' a little smaller than 1. This crossover with $p' < 1$ is however not sufficient to explain the observations of $p' = 0.93$ over 4 orders of magnitude in time $t_c - t$ (Fig. 1).

The failure time has a power-law singularity $\sim (s - s^*)^{-1/2}$ for $s \approx s^*$, as found previously [10] for the model with a linear dashpot, and decays exponentially for $s \gg s^*$ [8]. The transition time t_m (minima of the strain rate) is equal to $t_c/2$. This result recovers the proportionality of t_m and t_c found experimentally, but predicts a duration for the primary creep shorter than the observations $t_m \approx 2t_c/3$ (Fig. 3).

In conclusion, we have shown that the interactions between the RE elements together with a large heterogeneity and a simple nonlinear rheology is sufficient to explain qualitatively and quasi-quantitatively our experiments. This model replaces the need for complex memory effects (such as the integro-differential

Schapery long-memory formalism [12]) often invoked in the composite literature. A natural improvement of the model would be to relax the democratic load sharing rule as in [10] in order to introduce realistic elastic interactions. This improvement may provide a more realistic value of p' and of the constant of proportionality between t_m and t_c (Fig. 3).

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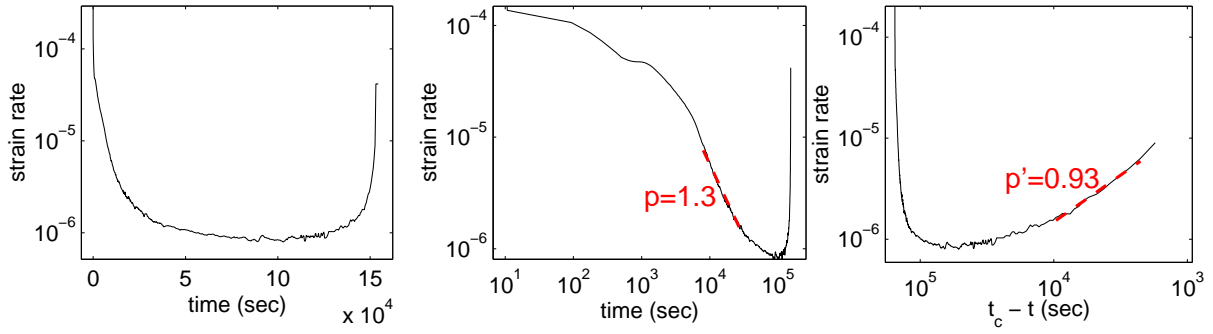


FIG. 1: Creep strain rate for a $[\pm 62^\circ]$ specimen. (a) linear time scale, (b) logarithmic time scale to test for the existence of Andrade's law in the primary creep, (c) logarithmic time scale in $t_c - t$ to test the time-to-failure power law in the tertiary creep.

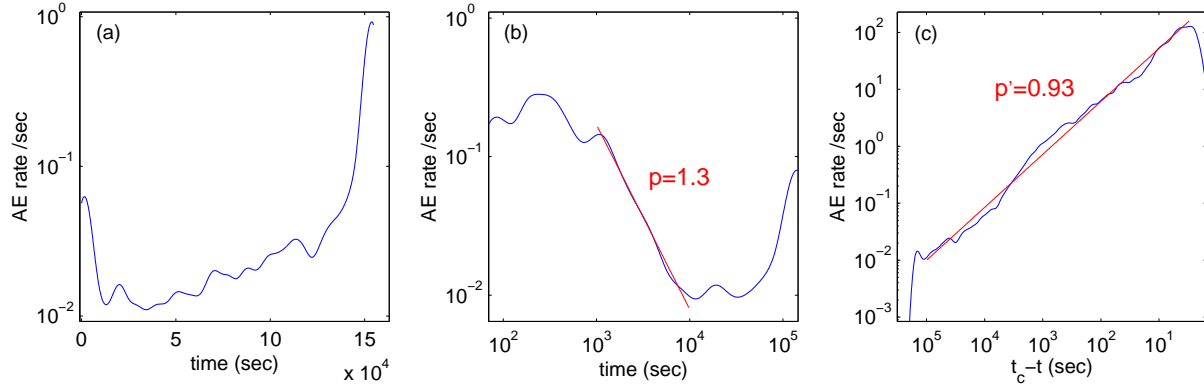


FIG. 2: Rate of AE events for a $[\pm 62^\circ]$ specimen. The three panels are as in Fig. 1.

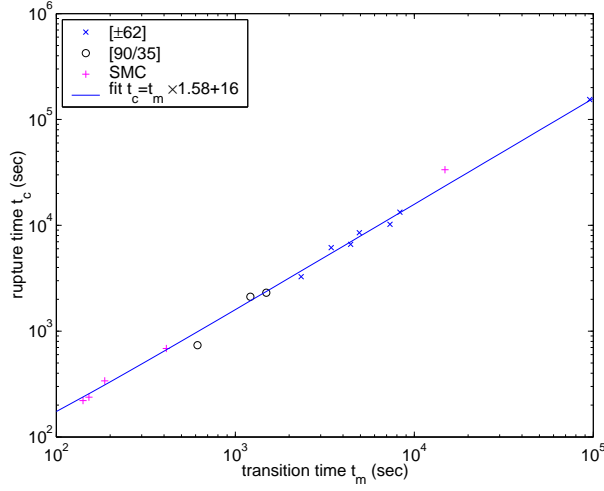


FIG. 3: Relation between the time t_m of the minima of the strain rate and the rupture time t_r , for all samples.

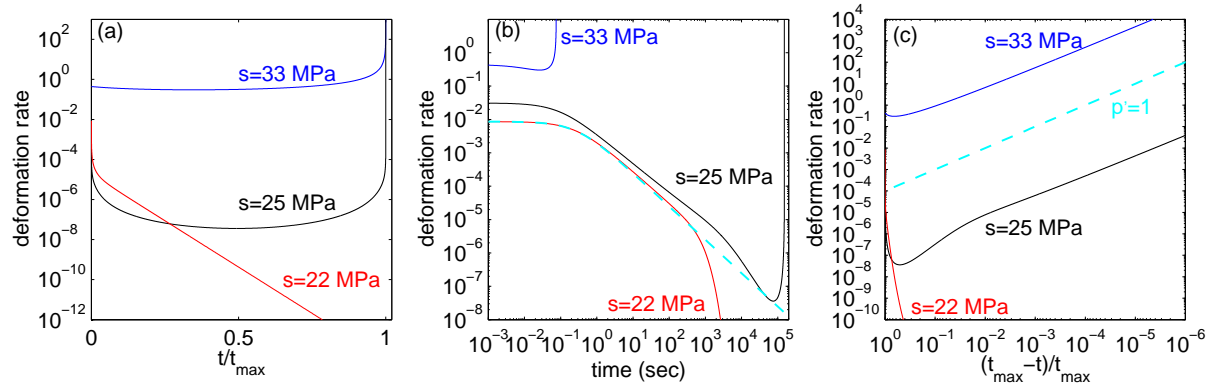


FIG. 4: Strain rate de/dt given by (1) for different values of the stress s , and with parameters $E = 20$ GPa, $\mu = 1.2$, $e_{01} = 0.003$, $e_{02} = 0.015$, $\beta = 50$ GPa $^{-1}$ and $K = 10^{-5}$ sec $^{-1}$. Panel (b) illustrates Andrade's law in the primary regime, with exponent $p \approx 1$ for $s = 22$ MPa and $p \approx 0.8$ for $s = 25$ MPa. The dashed line is the approximate solution (2) of (1) with $s = 22$ MPa. Panel (c) shows the power law acceleration of de/dt before failure for $s = 25$ MPa and $s = 33$ MPa, with $p' \approx 1$ asymptotically. In (a) and (c) the time is normalized by the rupture time for $s = 25$ MPa and for $s = 33$ MPa, and by the time when de/dt decreases below 10^{-14} for $s = 22$ MPa.